

CHARLES AUSTEN ANGELL

Biography

I was born in Canberra, Australia, in 1933, first child of Herbert and Kate Angell, whose memory I hold in enormous love and respect. My father was a plant pathologist with a wealth of hobbies ranging from model T Ford reassembling, through 6-in. reflecting telescope lens grinding, to stoneware pottery (fired in a kiln fueled by sump oil from the local garage—air pollution in Canberra in those days not being a grave problem). I think it was his casting of aluminum parts from scrap aircraft aluminum melted in the open-hearth dining-room fireplace that got me interested in hot liquids at an early age, and when it came time to choose undergraduate science courses at Melbourne University, I found the topics listed for chemical metallurgy far the most interesting.

After graduation, with Bachelor's and Master's degrees (but in fact specializing in field hockey which took me all around Australia), I went to work on molten salts with the electrochemist John Bockris at the University of Pennsylvania. This great experience introduced me to competitive science. I went on to Imperial College of Science in London as the Stanley Elmore Fellow in the Royal School of Mines to work with one of Bockris' first students, John Tomlinson, who was a master experimentalist. Since I had no Ph.D. at that time, I submitted the research done with Tomlinson on self-diffusion in molten Cd—CdCl₂ solutions and TiCl₃ for an Imperial College of Science Ph.D. and subsequently was awarded the biennial Armstrong Medal for graduate research at that institute. I owe it to I.C.'s effective safety procedures that, after working with radioactive thallium, an α -emitter, in the hot liquid state, I am still alive and well, forty years later!

I then took a position at Melbourne University and enjoyed Melbourne life for two years. There I mixed KNO₃ and Ca(NO₃)₂ with great skepticism after reading a Russian report that the liquid solution became glassy on cooling, and was astonished to find that it was true. How could one pull fibers from a melt of such simple particles without becoming convinced that the glassy state must be fundamental to understanding liquids? I was hooked on glassformers for life. I wrote my first single-author paper on this system, and have returned to it frequently in later years (most notably in the 1980's trans-Atlantic light scattering collaboration with Lena Torell and her group).

In 1964 the desire to be part of the American scientific scene became too strong and I took a leave and returned to the United States to work at Argonne National Laboratory with Dieter Gruen, who is still to this day doing something new every time I see him. He taught me spectroscopy and introduced me to Grant Urry from Purdue University who was scouting for new faculty. Purdue at that time had the second largest chemistry graduate student population in the country, and I changed my mind about return to Australia and tried out for the position, successfully. I was very lucky to be starting up then because Sputnik had just gone up and suddenly there was research money and many talented students interested in the scientific challenge.

With the help of students like Joe Wong and Ed Sare, post-doc K. J. Rao, and visiting faculty members Connie Moynihan and Alan Easteal, we characterized aqueous and molten salt glasses spectroscopically from the far-IR down through the glass

transition, and in 1970 started to ask troublesome thermodynamic questions about glassy water and its relation to the supercooled liquid, about which little was then known. Joe Tucker (with whom I subsequently lost contact for twenty years till this winter) measured the heat capacity of supercooled water, and found it made the Angell—Sare problem worse. Then Robin Speedy, as a post-doc from New Zealand, measured the compressibility and showed it and many other properties followed power laws with 228 K as a singular temperature. The paper took a while to write and we had to fight to get it published—and in spite of clever studies by many workers the meaning of it all is still controversial twenty years later, the sign of a good problem. (Our latest ideas just came out in *Nature*, April, 1999. More data of high quality were provided by my first co-workers from Japan, Hitoshi Kanno and Masaharu Oguni, who remain firm friends to this day. At this time Joe Wong and I finally finished a five-year book writing project and published *Glass: Structure by Spectroscopy* with eleven chapters. I'll never forget the seven-day-a-week, all-summer effort to write Chapter 11 on "relaxation spectroscopy"—my responsibility—after Joe had finished all his (nor the extraordinary help I had from two little office-camping, coffee-carrying daughters).

Thanks to Graham Hills of the University of Southampton and his NATO project with Earnest Yaeger, in 1972 I teamed up with Les Woodcock and Julian Clarke, who were starting up in his department. It was at one of this NATO group's mini meetings that I first gave a talk showing the T_g -scaled viscosity Arrhenius plot that was later to become well known. I still remember vitresearch Ted Litovitz' very positive reaction to this plot, but it took years before I found a vehicle for its publication (Martin Goldstein's first New York Academy of Sciences workshop on glassforming systems (1976)). Les had just pioneered the application of molecular dynamics to ionic systems and, under NATO auspices and with the help of Phil Cheeseman (then a Purdue undergraduate and assembly programmer), we extended his programs to the case of SiO₂. The project was initially somewhat tongue-in-cheek (ionic silica!), and we surprised ourselves and others with the nice results—which I think may have helped start a field in geochemical simulations. Les studied hard spheres, Julian studied LJ, and in 1980, under our own NATO grant, we argued our way through to the first review of applications of computer simulations to glassforming systems.

Stimulated, or perhaps pushed, by the Purdue University ARPA program (later NSF-MRL) I started to work on superionic glasses about this time and this was later to become a major component of our research. Also as a result of a 1979 sabbatical leave in Grenoble (Institute Laue-Langevin) I became involved in nucleation and growth of crystals from supercooled liquids. In this area I was greatly helped, not to say taught, by Doug Macfarlane, who came from New Zealand and mastered the field and his Ph.D. in a record 2.75 years including six months in France with Jacques Lucas doing fluoride glasses! Doug also fleetingly discovered the first glass-forming microemulsions, which are still to be fully exploited. In 1983 Jean-Louis Souquet and Dennis Ravaine organized one of the great conferences of

the decade in Grenoble—on Solid State Ionics—and I introduced Moynihan's decoupling index to the field. The mobile ion “glass transition” is still waiting for its moment in the sun.

In 1986 Kai Ngai organized in Blacksburg, VA, the first of what was later to become the triennial Kai Ngai pilgrimage on relaxation phenomena (Heraklion in 1991, Alicante in 1994, and Vigo in 1997). It was a small meeting but for one reason or another the T_g -scaled Arrhenius plot of viscosities, now dressed in the words “strong and fragile liquids” and bolstered with additional data made possible by emulsion studies, suddenly caught on and the conference proceedings paper became my best-known work. I have often wondered what factors are involved in the successful launching of a scientific idea, and have also often thought that this particular one may have got more attention than it merited, though it has undoubtedly been useful.

Fragile liquids have continued to haunt me. Familiar glassformers like silicates start strong at the silica end and become fragile as second components break up the low-density structure. But Hiro Tatsumisago and Jenny Green, who shared in the move from Purdue to Arizona State University in 1989, showed (in our first work at ASU in collaboration with friend and colleague Stuart Lindsay) that in covalent glasses Ge–As–Se one finds strong liquid behavior in mid-system at the Phillips–Thorpe magic average bond density of 2.4 bonds per particle—and at a high-density extremum. When *that* is properly understood we might understand what fragility is all about. Then Roland Böhmer correlated fragility with non-exponentiality, and also noted that Ian Hodge had correlated non-linearity with fragility—so all the canonical properties of the extended liquid state would seem to have a common origin. What can it be? Strong liquids are boring but the extreme of fragility is a first-order transition from liquid to glass, and *that* is worth chasing, particularly as it may provide a route to the ideal glass state, a state with little residual entropy. I think it might happen at the Ge-rich end of the Ge–As–Se system and indeed in many strongly over-constrained (tetrahedrally bonded) systems, best case, silicon. This may be why amorphous Si shows no typical “glassy anomalies” (Debye T^3 law violations). Neither does glassy water which I see as a related but borderline case.

ASU has been a fun place to work. People collaborate well here, and they enjoy their joint achievements. ASU geochemists

understand about fluid inclusions, and with advice and samples from John Holloway and, with George Wolf's expertise in microRaman spectroscopy, Jenny Green observed aqueous solutions at more than kilobar negative pressures. Thanks to student Qing Zheng, this became a systematic extension of the domain of metastable superheated and stretched water from the critical point to the homogeneous cavitation limit at ambient temperature. Much remains to be done here, with crystallization of guest-free water clathrates (Ice XVII?) a plum waiting to be picked. With Paul McMillan, John Holloway, Mike O'Keeffe, super post-doc Tor Grande, and *pressure*, we made purely nitride glasses, and Kurt Leinenweber is currently “pressure-tuning” the relative energies of liquid versus crystal to create a series of “boutique glassformers” of which quite simple substances like $\text{Ca}(\text{OH})_2$ and $\text{Sr}(\text{OH})_2$ are early examples. Finally, a number of us, including super post-doc Peter Poole, have been actively involved in the subject covered by George Wolf's term “polyamorphism”, which deals with the “vitreous polymorphs” that Ed Sare and I “footnoted” about so long ago. The connection to mesoscopic polyamorphism, as the folding of polypeptide chains into life-controlling enzymes and other proteins can be considered, intrigues me extremely and I hope to spend more time on it in the future.

Recently some practical benefits have emerged. With Changle Liu and Kang Xu, there have been patents on “polymer-in-salt” electrolytes, with Shengshui Zhang and Kang Xu there have been patents on quasi ionic and molecular solvent-based electrolytes, and with Hema Senapati and Vesselin Velikov there are patents in process on industrial refrigeration fluids. The latter turn out to be just the hot version of the glassforming lithium salt hydrates we worked on so long ago. Except now there are some new twists which I think will turn out to be important in a number of areas including electrolytes.

New things turn up all the time. How can we be so fortunate to work in a business like this? It has been and continues to be a very exciting life and I feel so deeply privileged to have been able to share it with so many fine, intelligent, and unbelievably enjoyable colleagues both in this country and around the world. To all of you, only a few of whom I have found ways to mention here, I say a heartfelt “Thank you!” and “Let there be more”.

Colleagues of C. Austen Angell

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